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Plastic carrier polishing chamber reduces pollution swapping from denitrifying woodchip bioreactors

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HIGHLIGHTS:

- Woodchip bioreactor columns were followed with plastic carrier polishing chambers.
- The polishing chambers did not enhance nitrate removal.
- The polishing chambers helped reduce bioreactor nitrite releases.
- A full-scale post-bioreactor polishing chamber may not be worth the added cost.

Abstract

Denitrifying bioreactors with solid organic carbon sources (i.e., “woodchip bioreactors”) have proven to be relatively simple and cost effective treatment systems for nitrate-laden agricultural and aquacultural waters and wastewaters. However, because this technology is still relatively new, design modifications, such as the addition of a post-bioreactor polishing chamber filled

with inert media, may offer potential to increase nitrate removal and mitigate unintended bioreactor by-products. Paired-column configurations filled with woodchips followed by plastic biofilm carrier media showed significant nitrate removal within the woodchip bioreactor columns (37, 26, and 88% nitrate removal efficiencies at woodchip column retention times of 7.1, 18, and 52 h), but no significant additional nitrate removal benefit of the post-processing plastic media chamber (41, 22, and 89% nitrate removal efficiencies, respectively). Releases of chemical oxygen demand from the woodchips were likely not sufficient to fuel significant nitrate removal in the polishing chamber. However, the polishing chamber significantly reduced nitrite releases from the bioreactor columns, and provided some mitigation of reduced sulfate during the 52-h retention time testing period (influent, woodchip effluent, and plastic chamber effluent sulfate concentrations of 23.6, 18.8, and 20.7 mg SO₄²⁻ L⁻¹, respectively). A full-scale post-woodchip polishing chamber filled with inert plastic media generally may not be worth the added cost unless the receiving waters are particularly sensitive to nitrite or hydrogen sulfide.

KEYWORDS: woodchip; denitrification; plastic carriers; wastewater; nitrate;

1. Introduction

Heterotrophic denitrifying bioreactors that use readily available solid carbon sources (e.g., woodchips) to provide relatively inexpensive treatment of nitrate (NO₃⁻) for agricultural and aquacultural effluents have gained much attention within the past decade (Addy et al., 2016; Lepine et al., 2016; von Ahnen et al., 2016). Such cost-competitive “woodchip bioreactor” denitrification technologies are simple solutions for these industries to meet increasingly stringent guidelines to reduce nitrogen pollution. However, while denitrifying woodchip

bioreactors have moved beyond the proof of concept (Christianson and Schipper, 2016), the newness of this technology merits investigation into more advanced designs that improve NO_3^- removal and/or minimize nitrite (NO_2^-) or hydrogen sulfide (H_2S) production under a variety of conditions while still maintaining the simplicity of the concept of a “trench filled with woodchips”.

Addition of inert media to the woodchips may help either maintain hydraulic conductivity of the bed over time (e.g., gravel/woodchip mixtures: Burbery et al., 2014; Herbert et al., 2014; Wildman, 2001) or serve as additional surface area for biological processes. Commercial plastic carriers for denitrifying moving bed reactors are specifically designed to maximize surface area for biofilm growth while maintaining void space for hydraulic conductivity, and the use of such engineered carriers is widely documented for attached growth biological treatment of wastewater (Tchobanoglous et al., 2003). Early work from Saliling et al. (2007) compared the use of woodchips, wheat straw, and plastic carriers for denitrification treatment of aquaculture wastewater using methanol as the carbon source, but it is now established that woodchips themselves are sufficient to fuel this process. Feyereisen et al. (2017) investigated the addition of plastic carriers following a column of corncob-fueled denitrification, and observed a 21% nitrate-removal improvement provided by the addition of the plastic carrier polishing chamber. Corncobs are a more labile carbon source than woodchips for denitrifying bioreactors, but are less commonly used for this application.

The objective of this column study was to evaluate potential nitrate-removal and pollution swapping (i.e., removal of NO_3^- at the expense of releases of other compounds) of a post-

woodchip bioreactor polishing chamber filled with inert plastic media. It was hypothesized that chemical oxygen demand (COD) released from the woodchips, during longer-term testing, would be sufficient to serve as a carbon source to fuel additional denitrification within the plastic carrier column.

2. Methods and materials

Two paired up-flow configurations each included a denitrification column (schedule 40 PVC; 15.2 cm diameter; 63 cm length; 11.1 L) packed with woodchips that fed a second column of the same dimensions filled with plastic carriers (WMT MB3 media with surface area $600 \text{ m}^2/\text{m}^3$; Baton Rouge, LA) ($n = 2$). The columns have previously been described by Christianson et al. (2017) who investigated pairing denitrifying woodchip bioreactors and P-sorbing filters; this polishing chamber evaluation was performed in conjunction with the previous experiment as an additional treatment. Locally obtained woodchips (Lowe's Products, Shepherdstown, WV, USA; a "hardwood blend") were sieved with coarse mesh, and the remaining woodchips had a D_{50} (i.e., median particle diameter) of approximately 6 mm with a 72% total porosity (particle size distribution with sieves of 13, 6.4, 5.6, 4.0, 2.0, 0.85, 0.60 mm, and pan: 5.5, 40, 13, 18, 17, 4.3, 0.8, and 1.6% by weight, respectively). The woodchip columns were packed in four layers (following ASTM, 2012) to an average bulk density of approximately 270 kg m^{-3} (woodchip dry weight of $3.0 \pm 0.20 \text{ kg}$). The plastic carriers (white polyethylene; 1.9 cm diameter; 1.6 cm height; $\approx 1.0 \text{ g}$ per piece; surface area $600 \text{ m}^2/\text{m}^3$; 85% void space; MB3 Media, Water Management Technologies, Baton Rouge, Louisiana) were loosely filled into the columns (i.e., not tamped). The woodchip columns were leached at a hydraulic retention time (HRT) of 6-8 h for 33 d prior

to starting the experiment (i.e., 33 d prior to $t = 0$ d) to help minimize impacts of start-up organics flushing on experimental results.

The experiment was fed with water discharged from the bottom drains of fish culture tanks that was treated with a microscreen drum filter. The water was dosed to achieve 27.5 ± 4.5 mg $\text{NO}_3\text{-N}$ L^{-1} (mean \pm standard deviation; range: 21.0-40.5 mg $\text{NO}_3\text{-N}$ L^{-1} ; $n = 56$ over the entire test) to be representative of both aquacultural effluents and agricultural tile drainage water, the most common application for woodchip bioreactors (aquacultural effluents ≈ 0 -400 mg $\text{NO}_3\text{-N}$ L^{-1} : Timmons and Ebeling (2010); tile drainage ≈ 10 -30 mg $\text{NO}_3\text{-N}$ L^{-1} : Jaynes et al. (1999)). The automated dosing tank which fed the columns operated in batch-mode (refilling between four times per day to every other day depending on the column flow rate) by pulling from continuously flowing fish tank discharge water; the columns were continuously fed from the dosing tank using a peristaltic pump (Masterflex L/S economy drive, Model: 7554-90). The paired configurations were operated under a range of retention times to represent three realistic conditions a bioreactor may experience: (1) initial operation under a low retention time simulating a spring tile drainage bioreactor installation (7.1 ± 1.6 h HRT; d 0-97); (2) a steady state period with a moderate retention time (18 ± 2.8 h HRT; d 217-287); and (3) an overly long retention time to study nitrate-limited conditions (52 ± 9.6 h HRT; d 301-336). The corresponding additional HRTs provided by the plastic media chambers were slightly longer due to the greater porosity of the plastic media versus the woodchips (85 vs. 72%, respectively; 8.1 ± 1.7 , 20.7 ± 3.3 , and 61.3 ± 11.4 h for the three operational phases, respectively). The 336 d test was conducted in a temperature-controlled greenhouse (influent water temperature: $17.6 \pm 2.0^\circ\text{C}$).

Water grab samples were collected weekly from the influent, effluent, and between the woodchip and polishing chambers, and were analyzed on-site for COD, nitrate-nitrogen (NO_3^- -N), nitrite-nitrogen (NO_2^- -N), sulfate (SO_4^{2-}), alkalinity, and pH following standard methods (APHA, 2005; Hach, 2003; Hach spectrophotometers DR 2700, DR 4000, and DR 6000, Loveland, CO, USA). Dissolved oxygen (DO) at these three locations was measured twice weekly (Hach HQ 40-d meter with LDO101-10) by collecting at least 150 mL of sample into insulated glass beakers to minimize water surface air interactions. Nutrient removal efficiencies were calculated by dividing the difference of the inflow and outflow loads by the inflow load either across only the woodchip column or across both the woodchip + plastic carrier columns. Nitrate-N removal rates ($\text{g N removed m}^{-3} \text{ d}^{-1}$) were calculated from the cumulative mass removed between two sample dates divided by the gross column volume (woodchip column 11.1 L; plastic carrier column 11.1 L) and the difference between the two sample dates. Differences between treatments were assessed using One-Way Analysis of Variance testing (Shapiro-Wilk normality test; Brown-Forsythe equal variance test; $\alpha = 0.05$); when data did not satisfy normality testing, a Kruskal-Wallis One-Way Analysis of Variance based on ranks was used.

3. Results and discussion

3.1 Nitrate removal

The addition of the plastic carrier polishing chamber improved NO_3^- removal efficiency from 37% (woodchip alone) to 41% (paired configuration) during the initial operation period, but this was not a statistically significant increase based on the effluent NO_3^- concentrations (Table 1; woodchip and plastic carrier effluent: 22.4 ± 6.4 and $19.8 \pm 6.0 \text{ mg NO}_3\text{-N L}^{-1}$, respectively). Similarly, there was no statistically significant difference between the NO_3^- concentrations

exiting the woodchip chamber and the polishing chamber during the steady state (18.1 ± 2.8 and 19.1 ± 2.7 mg $\text{NO}_3\text{-N L}^{-1}$, respectively; Table 2) or the overly long retention time periods (3.1 ± 3.1 and 2.7 ± 2.6 mg $\text{NO}_3\text{-N L}^{-1}$, respectively; Table 3). The significant decrease in NO_3^- concentrations across the woodchip columns alone compared to the influent for all periods, paired with the consistent increase in alkalinity across these columns (Tables 1-3), was a strong indication that NO_3^- removal was due to heterotrophic denitrification (van Rijn et al., 2006).

It was hypothesized that COD released from the woodchips, particularly during the initial operation and overly long retention time periods, would provide additional respiration substrate for denitrification within the plastic carrier column. Optimum COD: $\text{NO}_3\text{-N}$ ratios for complete N removal using heterotrophic denitrification with the use of readily available carbon sources range from 3:1 to 6:1 (van Rijn et al., 2006). During the initial and steady state phases, the 22.4 and 18.1 mg $\text{NO}_3\text{-N L}^{-1}$ exiting the woodchip columns would have required at least 67.2 and 54.3 mg COD L^{-1} (i.e., at least three times the NO_3^- concentration) for complete NO_3^- removal. While COD concentrations significantly increased across the woodchip column during the initial operation period (Table 1; 5.0 to 14.7 mg COD L^{-1}), the woodchip effluent COD concentration was much too low to facilitate complete NO_3^- removal especially considering only a portion of this COD would be biologically available. Feyereisen et al. (2017) observed 21% greater NO_3^- removal when a plastic carrier chamber was placed downstream of a corncob bioreactor (i.e., 44% N load reduction from the corncobs versus 54% N load reduction from the corn cob + plastic carrier treatment). However, corncobs are a relatively more labile carbon source compared to woodchips. Feyereisen et al.'s (2017) plastic carrier chamber removed approximately 6.92 ± 4.38 mg $\text{NO}_3\text{-N L}^{-1}$ above that provided by the corncobs at 15.5°C (based

on data in Supplemental Materials). They reported TOC concentrations exiting the corncobs of approximately 20-40 mg TOC L⁻¹ (Feyereisen et al., 2016), or approximately 60-120 mg COD L⁻¹ eluting from the corncobs assuming a general wastewater correlation of COD being approximately three times TOC (Dubber and Gray, 2010). This estimated corncob-fueled COD range entering the plastic carrier chamber aligns more closely with the COD:NO₃-N ratios required to facilitate additional denitrification than that observed here (i.e., 6.92 mg N L⁻¹ removed per an estimated 60-120 mg COD L⁻¹ supplied).

The 37% NO₃⁻ removal efficiency from the woodchip column at a 7.1 h HRT was greater than expected compared to the 26% removal efficiency during the steady state period (HRT 18 h; (Tables 1 and 2; Fig. 1a), as a shorter retention time generally correlates to a lower removal efficiency under non N-limited conditions (Lepine et al., 2016). While some of this result could be attributed to unavoidable experimental variation due to treatment of the variable water chemistry coming from an operational aquaculture system, early flushing of readily available carbonaceous compounds (Fig. 1b) was thought to have supported the high removal efficiencies occurring within the woodchip chamber observed during this initial operation phase. During the initial operation period, NO₃-N removal rates across the woodchips columns averaged 32±0.6 g N removed m⁻³ woodchip volume d⁻¹ (paired chamber configuration: 18±2.0 g N per total m³ of media per d; N removal in the plastic carrier chamber: 3.6±4.7 g N per m³ of plastic media per d), which was much greater than the 4.7 g N m⁻³ d⁻¹ woodchip bioreactors tend to average (Addy et al., 2016). Removal rates were more aligned with expected values during the steady state period (woodchip column: 7.0±0.8 g N m⁻³ woodchips d⁻¹; paired configuration: 3.0±0.2 g N m⁻³ total configuration volume d⁻¹; plastic carrier chamber: -1.1±0.3 g N per m³ of plastic media per

d). Removal rates during the steady state and final periods corroborated the statistical analyses in Tables 2 and 3 with significant nitrate removal across the woodchip column and no additional significant benefit across the plastic media (final period: woodchip column: $10 \pm 0.8 \text{ g N m}^{-3} \text{ woodchips d}^{-1}$; paired configuration: $5.1 \pm 0.2 \text{ g N m}^{-3} \text{ total configuration volume d}^{-1}$; plastic carrier chamber: $0.1 \pm 0.3 \text{ g N per m}^3 \text{ of plastic media per d}$).

3.2 Pollution swapping: COD, sulfate, and nitrite

The COD concentration significantly increased across the woodchip columns during the initial operation period (influent solution: $5.0 \text{ mg COD L}^{-1}$; woodchip effluent: $14.7 \text{ mg COD L}^{-1}$; Table 1) and the final measurement period (5.4 to 25 mg COD L^{-1} ; Table 3; Fig. 1b, note log scale). The final period also exhibited the most notable change in solution pH from 8.05 in the influent to 7.45 exiting the woodchip column (Table 3). The highly reduced conditions in the woodchips may have precipitated fermentation and organic acid production (Mirzoyan et al., 2010) thus lowering the pH more notably than during the two other study periods. Regardless, during periods of COD release, the polishing chamber reduced the leached COD concentrations, though these changes were not significant, and the final paired-column effluent was still elevated above the influent values (Tables 1 and 3).

The only significant difference in sulfate concentrations between the three sampling locations occurred during the long retention time period where influent sulfate concentrations of $23.6 \text{ mg SO}_4^{2-} \text{ L}^{-1}$ were reduced to $18.8 \text{ mg SO}_4^{2-} \text{ L}^{-1}$ (Table 3; Fig. 1c). As with the woodchip-released COD, the plastic carrier polishing chamber provided some mitigation of the reduced sulfate (plastic carrier effluent: $20.7 \text{ mg SO}_4^{2-} \text{ L}^{-1}$; not a significant difference). While dissolved

hydrogen sulfide concentrations were not measured, the plastic media may have provided surface area for the oxidation of sulfide back to sulfate. During the long retention time period, as well as during the steady state period, DO levels increased across the plastic media chamber potentially corroborating the occurrence of conditions conducive to aerobic treatment (Table 2 and 3).

While a relatively shorter retention time would eliminate the production of sulfides and fermentation-associated COD releases, the production of NO_2^- under shorter retention times may be a pollution swapping trade-off (Fig. 1). When N removal was not complete during the first two monitoring periods, NO_2^- was produced (Fig. 1d). These results confirm those of Hua et al. (2016) who also reported that effluent NO_2^- concentrations increased with decreasing HRT. The relatively stable conditions during the steady state period showed a notable benefit to the plastic carrier chamber with a reduction of NO_2^- concentrations to that not significantly different from the influent (Table 2). Effluent from the polishing chamber still exhibited a 246% increase over the influent, but that was an improvement compared to the use of woodchips alone during the steady state phase. Comparison across measurement periods showed that incomplete NO_3^- removal, observed during all but the final month of testing, will result in some NO_2^- release. However, NO_2^- release was minimized when the woodchip bioreactor was paired with the additional reactor filled with plastic media. This finding may provide insight into the greater cumulative nitrate removal of a similar plastic-media filter after corn cobs observed in Feyereisen et al. (2017), a study in which nitrite specifically was not measured (i.e., $\text{NO}_3\text{-N} + \text{NO}_2\text{-N}$ analyzed colorimetrically via cadmium reduction). This study also reported reduced cumulative production of N_2O at 15.5°C for the treatment with a plastic-medium polishing

chamber, suggesting that the plastic media filled chamber resulted in a more complete denitrification process.

4. Conclusions

A post-woodchip polishing chamber filled with inert plastic media provided no additional NO_3^- removal benefit, and generally may not be worth the added cost. However, the addition of plastic carriers did significantly reduce releases of COD and NO_2^- from the woodchips, as well as provided opportunity for sulfate to re-oxidize after it had been reduced to sulfide during long retention times. Several of these processes internal to woodchip bioreactor systems merit further investigation in future research. At the field scale, an extra polishing chamber might not be necessary unless the receiving waters are particularly sensitive to NO_2^- or H_2S .

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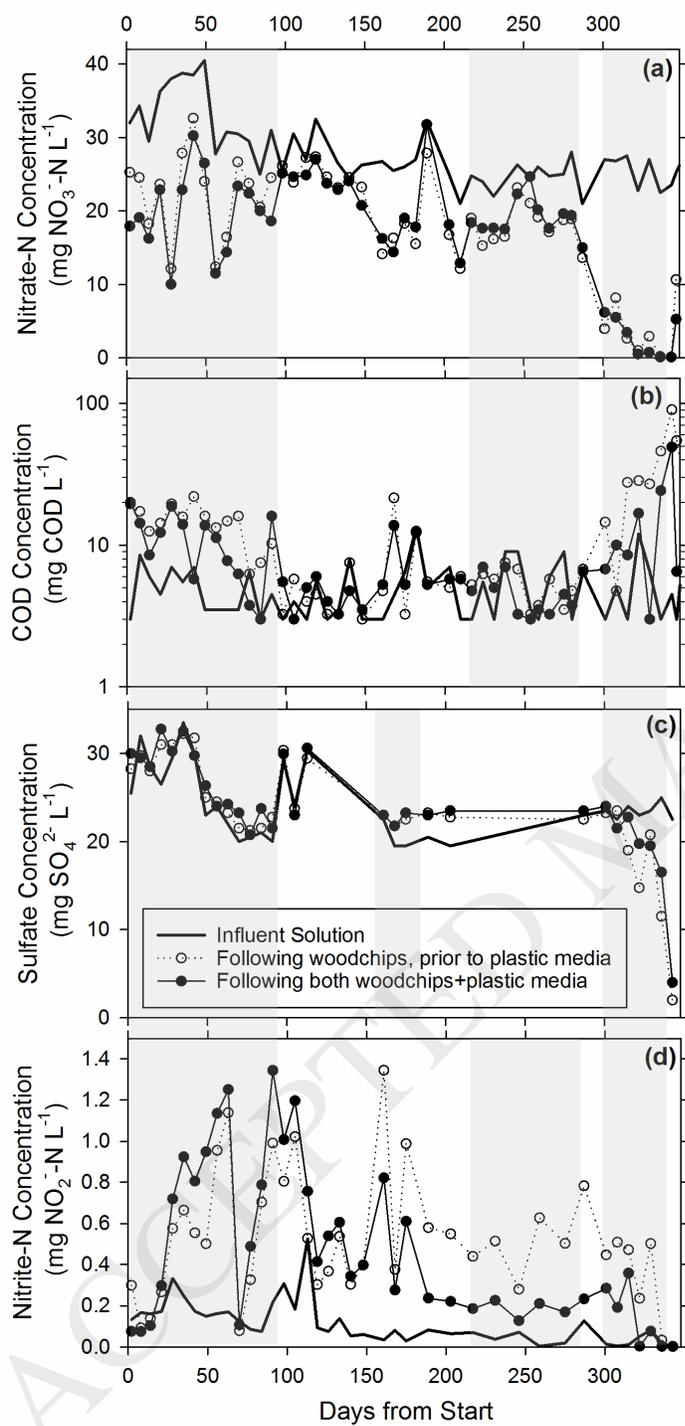
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Fig. 1. Mean nitrate (a), COD (b; log scale), sulfate (c), and nitrite (d) concentrations for the influent, woodchip effluent, and final effluent from the plastic carrier chambers ($n = 2$). The vertical gray shading represents the three analysis periods: (1) initial operation (7.1 h HRT; d 0-97), (2) steady state (18 h HRT; d 217-287 or d 161-175 for sulfate), and (3) overly long retention time (52 h HRT; d 301-336). Note, the steady state period used analysis dates of d 161-175 for sulfate due to reduced analytical capability in the lab.

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List of Tables**Table 1**

Initial operation period (d 0-97; 7.1 h hydraulic retention time in the woodchip column) mean \pm stdev nutrient concentrations for the influent, woodchip effluent and final effluent from the plastic carrier chamber. Negative removal efficiency indicates production of an analyte.

Concentration means followed by the same letter within a given row are not significantly different at $\alpha = 0.05$.

	Influent	Woodchip effluent	Plastic carrier effluent	Removal efficiency due to woodchips	Removal efficiency due to woodchips + plastic carrier
	----- mg L ⁻¹ -----			----- % -----	
Nitrate-N	33.0 \pm 4.7 a	22.4 \pm 6.4 b	19.8 \pm 6.0 b	37 \pm 1.4	41 \pm 3.9
COD	5.0 \pm 1.8 b	14.7 \pm 5.3 a	11.2 \pm 5.9 a	-165 \pm 14	-101 \pm 25
Sulfate ^a	25.4 \pm 4.6	25.7 \pm 6.2	26.9 \pm 4.0	2.5 \pm 0.6	-2.4 \pm 3.6
Nitrite-N	0.17 \pm 0.07 b	0.53 \pm 0.37 ab	0.67 \pm 0.46 a	-211 \pm 18	-300 \pm 85
pH	7.68 \pm 0.12 a	7.56 \pm 0.07 b	7.60 \pm 0.07 ab	1.5	1.0
Alkalinity	249 \pm 13 b	305 \pm 38 a	312 \pm 30 a	-23	-25
Dis. Oxygen	7.44 \pm 1.03 a	1.32 \pm 0.41 c	1.61 \pm 0.36 b	82	78

^a No statistically significant difference between the three sampling locations ($p = 0.515$)

Table 2

Steady state period (d 217-287; 18 h hydraulic retention time in the woodchip column) mean \pm stdev nutrient concentrations for the influent, woodchip effluent and final effluent from the plastic carrier chamber. Negative removal efficiency indicates production of an analyte. Concentration means followed by the same letter within a given row are not significantly different at $\alpha = 0.05$.

	Influent	Woodchip effluent	Plastic carrier effluent	Removal efficiency due to woodchips	Removal efficiency due to woodchips + plastic carrier
	----- mg L ⁻¹ -----			----- % -----	
Nitrate-N	24.6 \pm 1.9 a	18.1 \pm 2.8 b	19.1 \pm 2.7 b	26 \pm 3.7	22 \pm 2.3
COD ^a	5.5 \pm 2.6	5.4 \pm 1.7	4.7 \pm 1.7	0.3 \pm 1.5	13 \pm 7.0
Sulfate ^b	20.5 \pm 1.7	22.4 \pm 1.0	22.7 \pm 0.9	-7.9 \pm 2.9	-9.1 \pm 2.6
Nitrite-N	0.05 \pm 0.04 b	0.52 \pm 0.25 a	0.19 \pm 0.19 b	-825 \pm 374	-246 \pm 445
pH	7.82 \pm 0.13 a	7.60 \pm 0.08 b	7.62 \pm 0.08 b	2.9	2.5
Alkalinity	241 \pm 7 b	274 \pm 19 a	272 \pm 12 a	-14	-13
Dis. Oxygen	8.03 \pm 0.74 a	1.56 \pm 0.39 c	3.09 \pm 1.15 b	81	62

^a No statistically significant difference between the three sampling locations ($p = 0.420$)

^b No statistically significant difference between the three sampling locations ($p = 0.133$); analysis dates of d 161-175 due to reduced analytical capability in the lab ($n = 6$ due to three sampling events x two paired configurations)

Table 3

Long retention time period (d 301-336; 52 h hydraulic retention time in the woodchip column)
 mean \pm stdev nutrient concentrations for the influent, woodchip effluent and final effluent from
 the plastic carrier chamber. Negative removal efficiency indicates production of an analyte.
 Concentration means followed by the same letter within a given row are not significantly
 different at $\alpha = 0.05$.

	Influent	Woodchip effluent	Plastic carrier effluent	Removal efficiency due to woodchips	Removal efficiency due to woodchips plus plastic carrier
	----- mg L ⁻¹ -----			----- % -----	
Nitrate-N	25.6 \pm 2.3 a	3.1 \pm 3.1 b	2.7 \pm 2.6 b	88 \pm 2.8	89 \pm 0.3
COD	5.4 \pm 3.5 b	25 \pm 14 a	12 \pm 12 ab	-303 \pm 2.1	-94 \pm 119
Sulfate	23.6 \pm 0.9 a	18.8 \pm 4.7 b	20.7 \pm 2.8 ab	24 \pm 0.0	17 \pm 2.5
Nitrite-N	0.03 \pm 0.03 a	0.37 \pm 0.24 a	0.15 \pm 0.25 a	-1,130 \pm 291	-426 \pm 621
pH	8.05 \pm 0.12 a	7.45 \pm 0.05 b	7.51 \pm 0.08 b	7.4	6.7
Alkalinity	252 \pm 12 b	346 \pm 28 a	337 \pm 18 a	-37	-34
Dis. Oxygen	8.69 \pm 0.80 a	1.80 \pm 0.67 c	2.82 \pm 0.88 b	79	68